

# Vibrational Sidebands and Kondo-effect in Molecular Transistors

Jens Paaske<sup>1,2</sup> and Karsten Flensberg<sup>2</sup>

<sup>1</sup>*Institut für Theorie der Kondensierten Materie, Universität Karlsruhe, 76128 Karlsruhe, Germany*

<sup>2</sup>*Ørsted Laboratory, Niels Bohr Institute fAPG, Universitetsparken 5, 2100 Copenhagen, Denmark.*

(Dated: February 2, 2008)

Electron transport through molecular quantum dots coupled to a single vibrational mode is studied in the Kondo regime. We apply a generalized Schrieffer-Wolff transformation to determine the effective low-energy spin-spin-vibron-interaction. From this model we calculate the nonlinear conductance and find Kondo sidebands located at bias-voltages equal to multiples of the vibron frequency. Due to selection rules, the side-peaks are found to have strong gate-voltage dependences, which can be tested experimentally. In the limit of weak electron-vibron coupling, we employ a perturbative renormalization group scheme to calculate analytically the nonlinear conductance.

PACS numbers: 73.63.Kv, 73.23.Hk, 72.10.Fk, 72.15.Qm, 05.10.Cc

In recent years, the study of transport in mesoscopic systems has branched into investigations of single-electron devices based on single-molecule transistors [1, 2, 3, 4, 5]. Of particular interest is the possibility of combining electronics with mechanics, such that the vibrational or configurational modes of the molecule are coupled to its charge state. A number of interesting issues have already been addressed in this new field of nano-electro-mechanics. Firstly, it was shown by Park *et al.* [2] that quantum mechanical behavior of the center of mass oscillation of a C<sub>60</sub> can be excited by the tunneling electrons and a series of assisted steps were observed in the current. Similar structures have later been observed in a number of other experiments. These steps were associated with simple Franck-Condon physics, which means that the tunneling rates are modified by the overlaps of the initial and final states of the oscillator.

It is well established [6, 7] that *single particle* resonance tunneling is not destroyed by the electron-vibron coupling, but instead the resonance breaks up into a number of vibron sidebands. The question remains, though, whether more intricate *many-body* effects, such as the Kondo resonance, also cooperate with the electron-vibron coupling to form “Kondo sidebands”. The usual Kondo resonance has been observed in several molecular devices [3, 4] at unusually high temperatures, and recent experiments [5] on C<sub>60</sub>, and Co based transistors have revealed marked sideband resonances, which were suggested to arise from the interplay of a Kondo resonance with a vibrational mode.

In this letter we demonstrate that, in contrast to sequential tunneling, which is suppressed by the Franck-Condon overlap factors, the Kondo resonance remains intact well inside the Coulomb blockade valley. In fact, the electron-vibron coupling is predicted to enhance the exchange-coupling and thereby the Kondo-temperature. Maintaining the quantum coherence of vibrons, we show that the Kondo-resonance breaks up into a series of vibron sidebands. Moreover, we demonstrate that parity selection rules prohibit all sidebands at odd multiples of

the oscillator frequency, when tuning the gate-voltage to the particle-hole symmetric point.

Assuming the energy-level spacing on the molecule to be much larger than the charging-energy, the system may be described by the Anderson-Holstein Hamiltonian

$$H = \sum_{\alpha, \mathbf{k}, \sigma} \xi_{\alpha \mathbf{k}} c_{\alpha \mathbf{k} \sigma}^\dagger c_{\alpha \mathbf{k} \sigma} + \varepsilon_d n_d + \omega_0 b^\dagger b + U n_{d\uparrow} n_{d\downarrow} + \sum_{\alpha, \mathbf{k}, \sigma} (t_{\alpha \mathbf{k}} d_\sigma^\dagger c_{\alpha \mathbf{k} \sigma} + \text{h.c.}) + \lambda \omega_0 (b + b^\dagger) n_d. \quad (1)$$

where  $c_{\alpha \mathbf{k} \sigma}^\dagger$  and  $d_\sigma^\dagger$  are creation operators for electrons in the left and right conduction bands ( $\alpha = L, R$ ) and on the molecular quantum dot, respectively,  $n_{d\sigma} = d_\sigma^\dagger d_\sigma$ ,  $n_d = n_{d\uparrow} + n_{d\downarrow}$  and  $\xi_{\alpha \mathbf{k}} = \varepsilon_{\mathbf{k}} - \mu_\alpha$ . The vibrational mode of the molecule is created by the vibron operator  $b^\dagger$ , and  $\lambda$  denotes the dimensionless coupling strength. Describing the molecule as a quantum-dot, we have  $\varepsilon_d = (1 - 2N)E_C$  and  $U = 2E_C$ , in terms of the charging energy  $E_C$  and the mean occupation number  $N = C_g V_g / e$ , determined by the gate voltage,  $V_g$ , and the capacitance to the gate,  $C_g$ . Unless explicitly stated otherwise, we shall henceforth work in units where  $e = \hbar = k_B = 1$ .

Following Lang and Firsov [8], the electron-vibron coupling in the Hamiltonian (1) is eliminated by the unitary transformation  $H' = e^{iS_p} H e^{-iS_p}$ , with  $S_p = i\lambda(b - b^\dagger)n_d$ :

$$H' = \sum_{\alpha, \mathbf{k}, \sigma} \xi_{\alpha \mathbf{k}} c_{\alpha \mathbf{k} \sigma}^\dagger c_{\alpha \mathbf{k} \sigma} + \varepsilon'_d n_d + \omega_0 b^\dagger b + U' n_{d\uparrow} n_{d\downarrow} + \sum_{\alpha, \mathbf{k}, \sigma} (t_{\alpha \mathbf{k}} e^{\lambda(b^\dagger - b)} d_\sigma^\dagger c_{\alpha \mathbf{k} \sigma} + \text{h.c.}), \quad (2)$$

where  $\varepsilon'_d = \varepsilon_d - \lambda^2 \omega_0$  and  $U' = U - 2\lambda^2 \omega_0$ . We now consider the weak-tunneling limit,  $\Gamma_{\alpha \mathbf{k}} = 2\pi N(0) |t_{\alpha \mathbf{k}}|^2 \ll \min(-\varepsilon'_d, \varepsilon'_d + U')$ , where  $N(0)$  denotes the conduction electron (*ce*) density of states. In this limit, a generalized Schrieffer-Wolff transformation, devised by Schüttler and Fedro [9], may be used to eliminate all first order terms

in  $t_\alpha$ . To this end, we introduce the generator

$$S_v = i \sum_{\alpha, \mathbf{k}, \sigma, \eta} (t_{\alpha \mathbf{k}} \zeta_{\alpha \mathbf{k} \sigma \eta} n_{d\bar{\sigma}}^\eta d_{\sigma}^\dagger c_{\alpha \mathbf{k} \sigma} - \text{h.c.}), \quad (3)$$

where  $n_{d\bar{\sigma}}^\eta = (1 - \eta)/2 + \eta n_{d\bar{\sigma}}$ , with  $\eta = \pm 1, \bar{\sigma} = -\sigma$ , and  $\zeta_{\alpha \mathbf{k} \sigma \eta} = i \int_0^\infty dt e^{-i(E_{\alpha \mathbf{k} \eta} - i0_+)t} e^{-A(t)}$ , with  $E_{\alpha \mathbf{k} \eta} = \xi_{\alpha \mathbf{k}} - \varepsilon'_d - (1 + \eta)U/2$  and  $A(t) = \lambda(e^{-i\omega_0 t} b - e^{i\omega_0 t} b^\dagger)$ . Applying the transformation  $H'' = e^{iS_v} H' e^{-iS_v}$  and expanding to second order in  $t_\alpha$ , one finds that  $H'' = H'_0 + H_{\text{spin}} + H_{\text{dir}} + H_{\text{pair}}$ . We neglect the renormalization of the kinetic energy term in  $H'_0$  and, restricting to the regime of single occupancy, i.e.  $\mathcal{N} \approx 1$  and  $\lambda\omega_0 \ll E_C$  ( $U' > 0$ ),  $H_{\text{pair}}$  vanishes. The potential scattering term,  $H_{\text{dir}}$ , is omitted since it leads to no logarithmic singularities, and altogether we obtain the effective Hamiltonian

$$H'' = \sum_{\alpha, \mathbf{k}, \sigma} \xi_{\alpha \mathbf{k}} c_{\alpha \mathbf{k} \sigma}^\dagger c_{\alpha \mathbf{k} \sigma} + \omega_0 b^\dagger b + \sum_{\alpha, \mathbf{k}, \sigma; \alpha', \mathbf{k}', \sigma'} \mathbb{J}_{\alpha, \mathbf{k}; \alpha', \mathbf{k}'} \mathbf{S} \cdot c_{\alpha' \mathbf{k}' \sigma'}^\dagger \frac{\tau_{\sigma' \sigma}}{2} c_{\alpha \mathbf{k} \sigma}, \quad (4)$$

where  $\mathbf{S} = \frac{1}{2} d_{\sigma}^\dagger \boldsymbol{\tau}_{\sigma' \sigma} d_{\sigma}$  denotes the local spin-1/2, and  $\mathbb{J}_{\alpha, \mathbf{k}; \alpha', \mathbf{k}'} = t_{\alpha' \mathbf{k}'}^* t_{\alpha \mathbf{k}} [(X_{\alpha \mathbf{k}}^- - X_{\alpha \mathbf{k}}^+) + (X_{\alpha' \mathbf{k}'}^- - X_{\alpha' \mathbf{k}'}^+)]$  with  $X_{\alpha \mathbf{k}}^\eta = i \int_0^\infty dt e^{-i(E_{\alpha \mathbf{k} \eta} - i0_+)t + i\eta\lambda^2 \sin(\omega_0 t)} e^{A(0) - A(t)}$ .

In this effective Kondo-model, the exchange-coupling  $\mathbb{J}$  incorporates the dynamics of the vibron through the displacement operator  $e^A$ . In the vibron number-state basis it is therefore convenient to introduce Franck-Condon factors  $f_{n'n} = \langle n' | e^{-A(0)} | n \rangle$  [18], which allows us to write the matrix-elements of  $\mathbb{J}$  in the more transparent form:

$$J_{\alpha', \mathbf{k}'; \alpha, \mathbf{k}}^{n'n} \equiv \langle n' | \mathbb{J}_{\alpha', \mathbf{k}'; \alpha, \mathbf{k}} | n \rangle = t_{\alpha' \mathbf{k}'}^* t_{\alpha \mathbf{k}} \sum_{m=0}^\infty \left\{ f_{mn} f_{mn} \left[ \frac{1}{\xi_{\alpha \mathbf{k}} - \varepsilon_- + (m - n')\omega_0} + \frac{1}{\xi_{\alpha' \mathbf{k}'} - \varepsilon_- + (m - n)\omega_0} \right] - f_{n'm} f_{nm} \left[ \frac{1}{\xi_{\alpha \mathbf{k}} - \varepsilon_+ - (m - n)\omega_0} + \frac{1}{\xi_{\alpha' \mathbf{k}'} - \varepsilon_+ - (m - n')\omega_0} \right] \right\}, \quad (5)$$

valid for  $\xi_{\alpha \mathbf{k}}, \xi_{\alpha' \mathbf{k}'}, n\omega_0, n'\omega_0 \ll \min(\varepsilon_+, -\varepsilon_-)$ , where  $\varepsilon_- = \varepsilon'_d$  and  $\varepsilon_+ = \varepsilon'_d + U'$  are the energies of intermediate, empty, or doubly occupied states. In this sum, the energies of intermediate vibron states  $|m\rangle$  shift the energy denominators and the Franck-Condon factors determine the overlap between initial and final vibron states with intermediate states of the oscillator shifted by  $\sqrt{2}\lambda\ell_0$ , where  $\ell_0$  is the characteristic oscillator-length.

Since  $\sum_{m=0}^\infty f_{n'm} f_{nm} = \delta_{n'n}$  and  $f_{n'n} \rightarrow \delta_{n'n}$  for  $\lambda \rightarrow 0$ , the usual exchange-coupling,  $J_{\alpha' \alpha} = 4t_{\alpha'}^* t_{\alpha} / E_C$ , is recovered in either of the limits  $\omega_0 \rightarrow 0$  or  $\lambda \rightarrow 0$ . More generally,  $J^{n'n}$  may be represented as an

asymptotic power series as  $E_C/\omega_0 \rightarrow \infty$ , with leading terms  $J_{\alpha' \alpha}^{n'n} \propto J_{\alpha' \alpha} (\lambda\omega_0/E_C)^{|n'-n|}$ . In terms of the incomplete Gamma function,  $\gamma(\alpha, x)$ , one has  $J_{\alpha' \alpha}^{00} = J_{\alpha' \alpha} e^{-\lambda^2} (E_C/\omega_0) \sum_{\eta=\pm} (-\lambda^2)^{\eta\varepsilon_\eta/\omega_0} \gamma(\eta\varepsilon_\eta/\omega_0, -\lambda^2)$ , or simply  $J^{00} \approx J_{\alpha' \alpha} [1 + (\lambda\omega_0/E_C)^2]$  for  $\lambda\omega_0 \ll E_C$  and  $\mathcal{N} = 1$ , as found earlier in Ref. [9]. Staying well inside the Kondo-regime, any finite  $\lambda$  thus leads to a slight *enhancement* of  $J^{00}$ , and thereby of the associated Kondo temperature,  $T_K \sim D e^{-1/N(0)J^{00}}$  ( $2D$  being the *ce* bandwidth). This is essentially due to the vibron induced reduction of  $|\varepsilon_\pm|$ . Note also that the (cotunneling) amplitude  $J^{00}$  involves only *virtual* shifts of the oscillator and therefore no Franck-Condon overlap reduces its magnitude. This is in sharp contrast to the resonant (sequential) tunneling amplitude, relevant outside the Coulomb-blockade regime, which involves *real* excitations of the oscillator and is thereby reduced by a factor of  $|f_{00}|^2$  [2, 7].

We now consider the case of strongly asymmetric and momentum independent tunneling amplitudes,  $\Gamma_L \gg \Gamma_R$ . The current traversing the molecule from left to right is then given simply as [11]

$$I = -\frac{2e}{h} \Gamma_R \sum_\sigma \int d\varepsilon [f_L(\varepsilon) - f_R(\varepsilon)] \text{Im} \mathcal{G}_{\sigma\sigma}^R(\varepsilon). \quad (6)$$

From the equations of motion for the Hamiltonian (1), the local density of states is found to be related to the *ce* T-matrix as  $\text{Im}[\mathcal{G}_\sigma^{d,R}(\omega)] = |t_\alpha|^{-2} \text{Im}[\mathbb{T}_{\alpha\alpha}^{\sigma\sigma}(\omega)]$ , and the latter can now be obtained using the effective Hamiltonian (4). To third order in  $J_{\alpha, \alpha'}$ , we find that

$$\begin{aligned} \text{Im}[\mathbb{T}_{\alpha' \alpha}^{\sigma' \sigma}(\Omega)] &= -\delta_{\sigma' \sigma} \frac{3\pi}{16} N(0) (1 - e^{-\omega_0/T}) (1 + e^{-\Omega/T}) \\ &\times \sum_{\substack{lmn \\ \alpha_1 \alpha_2}} J_{\alpha' \alpha_1}^{nm} J_{\alpha_2 \alpha}^{ln} e^{-n\omega_0/T} [1 - f(\Omega + (n-l)\omega_0)] \\ &\times \theta(D - |\Omega + (n-l)\omega_0|) \left\{ \delta_{ml} \delta_{\alpha_1 \alpha_2} + N(0) J_{\alpha_1 \alpha_2}^{ml} \right. \\ &\times \left. \left[ \ln \left| \frac{D}{\Omega + (n-m)\omega_0} \right| + \ln \left| \frac{D}{\Omega + (m-l)\omega_0} \right| \right] \right\}, \quad (7) \end{aligned}$$

with the shorthand notation  $\ln|D/x| = \ln|D/\sqrt{x^2 + T^2}|$ . In the asymmetric limit considered here, we take  $\mu_L = 0$  and bias the right lead to  $\mu_R = -V$ , leaving the position of the molecular energy levels unaffected. For  $T \ll \omega_0 \ll D$ , the differential conductance is then obtained from Eq. (6) as  $G(V) = -\frac{2e^2}{h} (\Gamma_R/\Gamma_L) N(0) \sum_\sigma \text{Im}[\mathbb{T}_{LL}^{\sigma\sigma}(eV)]$ . From Eq. (7), the differential conductance appears to diverge as  $\ln(D/T)$  at voltages corresponding to multiples of the oscillator frequency, reflecting the onset of a Kondo-effect assisted by coherent vibron-exchange. In Fig. 1, the upper panel shows a gray-scale plot of  $\partial^2 I / \partial V^2$  as a function of bias-voltage  $V$  and mean occupation number (gate-voltage)  $\mathcal{N} = C_g V_g / e$ . The lower panel shows three cuts revealing the side-band resonances on the flanks of the central zero-bias resonance.

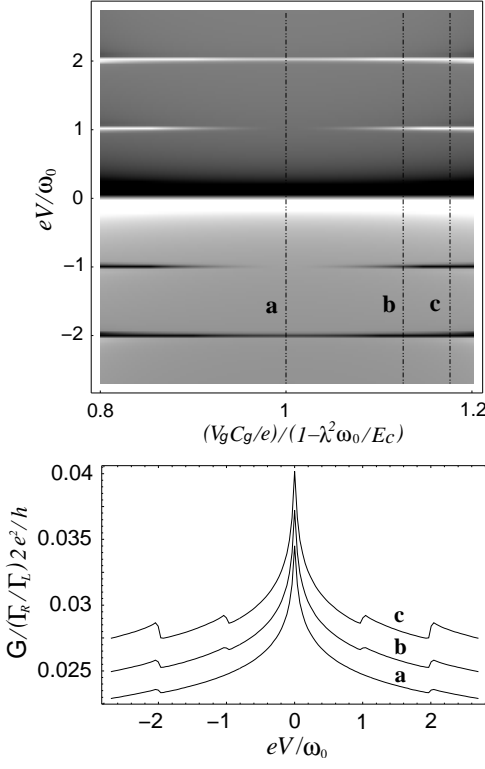


FIG. 1: Upper panel:  $\partial^2 I / \partial V^2$  vs. bias and gate voltage, for  $\lambda^2 = 3$ ,  $N(0)|t_L|^2 = 0.1\omega_0$ ,  $D = E_C = 8\omega_0$ , and  $T = 0.01\omega_0$ . Black/white indicates large negative/positive values. Lower panel: Conductance vs. bias voltage for three values of  $V_g$  corresponding to the vertical black lines (a,b,c) in the upper panel. The lower curve (a) corresponds to the ph-symmetric point  $\mathcal{N} = 1 - \lambda^2\omega_0/E_C$ .

By tuning the gate-voltage to  $\mathcal{N} = 1 - \lambda^2\omega_0/E_C$  one reaches the particle-hole (ph) symmetric point where  $\varepsilon_+ = -\varepsilon_-$ , and using the general symmetry  $f_{n'n} = (-1)^{|n'-n|}f_{nn'}$ , one finds from Eq. (5) that  $J_{n'n} \propto [1 + (-1)^{|n'-n|}]$ , implying that all spin-exchange processes involving the emission or absorption of an *odd* number of vibrons are prohibited at this particular gate-voltage. This parity selection rule, reflecting the inversion symmetry of the Kondo-Hamiltonian (4) at low energies, has important experimental bearings, since it predicts that all side-band resonances in the differential conductance located at voltages equal to *odd* multiples of  $\omega_0$ , must vanish when tuning the gate-voltage to the symmetric point, corresponding to the midpoint of the Coulomb-blockade valley. This is apparent in Fig. 1, where the conductance peak at  $V = \omega_0$  disappears as  $\mathcal{N}$  approaches the symmetric point (cf. curve a). Note, however, that any appreciable vibron modulation of the tunneling amplitudes will break the inversion symmetry[10] and thereby destroy this selection rule.

The logarithmic divergences appearing in third order perturbation theory call for a resummation of lead-

ing logarithmic contributions to all orders. This is done using the perturbative renormalization group (RG) method for frequency dependent couplings developed in Ref. [12]. Parametrizing the dimensionless couplings,  $g_{n'n} = N(0)J_{LL}^{n'n}$ , by the total energy of the ingoing conduction electron and vibron-state, we arrive at the infinite hierarchy of coupled (1-loop) RG-equations:

$$\frac{\partial g_{n'n}(\omega)}{\partial \ln D} = -\frac{1}{2} \sum_{m=0}^{\infty} [g_{n'm}(0)g_{mn}((m-n)\omega_0)\Theta_{\omega+(n-m)\omega_0} + g_{n'm}((n'-m)\omega_0)g_{mn}(0)\Theta_{\omega+(m-n')\omega_0}], \quad (8)$$

with  $\Theta_{\omega} = \Theta(D - |\omega|)$ . We shall restrict our attention to the ph-symmetric point and assuming that  $\lambda\omega_0 \ll E_C$  we may truncate this hierarchy and consider merely the lowest four coupled equations involving  $g_{00}$ ,  $g_{02} = g_{20}$  and  $g_{22}$ . The solution to this reduced set of equations is characterized by the parameters

$$\delta = \frac{g_{22} - g_{00}}{g_{00}g_{22} - g_{20}^2}, \quad \alpha = \frac{2g_{20}}{g_{00}g_{22} - g_{20}^2} \quad (9)$$

and  $T_K = D e^{-2/(g_{00}+g_{22}+\sqrt{(g_{22}-g_{00})^2+4g_{20}^2})}$ , where  $g_{n'n} \equiv g_{n'n}(D; 2n\omega_0)$ . All three parameters are invariant under the perturbative RG-flow from the initial cut-off  $D_0$  down to  $D = 2\omega_0$ . At scale  $D_0$ , we have  $\alpha, \delta \sim (\lambda\omega_0/E_C)^2/g_{00}$ , and therefore our truncation of Eq. (8) remains valid throughout the RG-flow roughly when  $\max(\alpha, \delta)/\ln(T/T^*) \ll 1$  (see below). Staying within the perturbative regime, we assume that  $\omega_0 \gg T \gg T_K$ .

We first solve the RG-eqs. for the constant coefficients  $g_{n'n}$  and the frequency dependent renormalized couplings are then obtained simply by integrating Eqs. (8):

$$g_{00}(\omega) = \frac{1}{\ln(|\omega|/T^*)} + \frac{\alpha^2}{8 \ln^2(2\omega_0/T^*)} \quad (10)$$

$$\times \sum_{\nu=0,1} \Theta_{\nu}(\omega) \left( \frac{1}{\ln(|\omega - 2\nu\omega_0|/T^*)} - \frac{1}{\ln(2\omega_0/T^*)} \right) \\ g_{20}(\omega) = \frac{\alpha}{4} \sum_{\nu=0,1} \left[ \frac{1}{\ln^2(\max(2\omega_0, |\omega - 2\nu\omega_0|)/T^*)} \right. \\ \left. + \frac{2\Theta_{\nu}(\omega)}{\ln(2\omega_0/T^*)} \left( \frac{1}{\ln(|\omega - 2\nu\omega_0|/T^*)} - \frac{1}{\ln(2\omega_0/T^*)} \right) \right], \quad (11)$$

with  $T^* = T_K(T_K/\omega_0)^{(\sqrt{\alpha^2+\delta^2}+\delta)/(2\ln(2\omega_0/T_K)+\sqrt{\alpha^2+\delta^2}-\delta)}$  and  $\Theta_{\nu}(\omega) = \Theta(2\omega_0 - |\omega - 2\nu\omega_0|)$ . Note that in Eqs. (10,11) we have retained only terms which will contribute to order  $\max(\alpha, \delta)^2/\ln^2(T/T^*)$  in the conductance.

While the logarithmic singularities at  $\omega = 0$  are cut off by temperature, those at  $\omega = 2\omega_0$  will instead be contained by  $\sqrt{T^2 + \gamma^2}$ , with  $\gamma$  given by the transition rate from vibron-state  $|2\rangle$  to  $|0\rangle$ . Using the golden rule with the renormalized coupling  $g_{20}(\omega)$ , we find  $\gamma \approx \pi\omega_0\alpha^2/(4\ln^2(2\omega_0/T_K))$ . Similarly, the broadening of the vibron states, induces a broadening of the step-functions

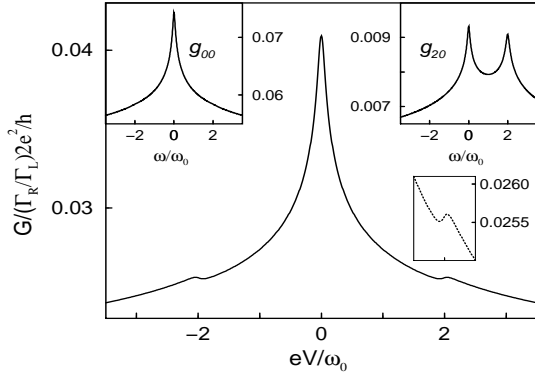


FIG. 2: Conductance versus bias voltage using *perturbatively* renormalized couplings. Gate voltage is tuned to the *ph*-symmetric point and  $\alpha = 4.30$ ,  $\delta = 7.54$ ,  $T_K = 1.51 \times 10^{-4} \omega_0$ ,  $T^* = 7.18 \times 10^{-8} \omega_0$ , and  $T = 0.05 \omega_0$ , i.e.  $\alpha / \ln(T/T^*) = 0.32$  and  $\delta / \ln(T/T^*) = 0.56$ , corresponding to bare parameters:  $\lambda^2 = 4.5$ ,  $N(0)|t_L|^2 = 0.1 \omega_0$  and  $D = E_C = 8 \omega_0$ . Insets show the renormalized couplings  $g_{00}(\omega)$  and  $g_{20}(\omega)$ , as well as a zoom in the conductance curve showing the satellite peak on a separate conductance scale but on same voltage scale.

in Eqs. (10,11) by  $T$  or  $\sqrt{T^2 + \gamma^2}$  for steps near  $\omega = 0$  or  $\omega = 2\omega_0$ , respectively.

The renormalized conductance is now obtained by inserting in the formula  $G(V) = (2e^2/h)(\Gamma_R/\Gamma_L)(3\pi^2/4)[g_{00}(eV)^2 + \sum_{\nu=\pm} \Theta(\nu eV - 2\omega_0)g_{20}(\nu eV)^2]$ , and indeed when expanding this result in bare couplings, we recover the result obtained by expanding Eq. (7) to order  $(\lambda\omega_0/E_C)^4$ . Including the broadening in both couplings and  $\Theta$ , the result is plotted in Fig. 2. This figure bears a certain resemblance to the curve obtained in Ref. [13] at  $T \ll T_K$ , using an entirely different method.

The possible decoherence effects which will arise when coupling the oscillator to phonons within the leads remain an open question. It is straightforward to generalize the Schrieffer-Wolff transformation applied here to a system where the molecule-oscillator is coupled to a separate bath of oscillators. However, even determining the effects on the leading logarithms for a given Q-factor involves a rather involved cumulant expansion. We expect the Kondo-effect to be more pronounced when dealing solely with *intra*-molecular vibrations, since these have been demonstrated to have particularly large Q-factors [14].

In the case of nearly symmetric couplings, we can no longer assume the oscillator to be in equilibrium with the conduction electrons of one specific side of the junction[15]. In line with the findings of Ref. [13, 16], we expect that nonequilibrium effects may in fact serve to enhance the Kondo side-peaks.

In conclusion, we have demonstrated the viability of an inelastic Kondo-effect carried by coherent vibron-assisted exchange tunneling, which can be observed as Kondo-sidebands in the nonlinear conductance. In contrast to the case of an applied microwave field [17], the zero-bias resonance is *not* suppressed by the vibronic coupling, and

it may therefore be difficult to discern the satellites from the background conductance. Nevertheless, even with very weak satellites, it should be possible to track their dependence on  $V_g$  (possibly in a plot of  $\partial^2 I / \partial V^2$ ) and thereby test our prediction that satellites at odd multiples of  $\omega_0$  are strongly reduced at the *ph*-symmetric point. Faint sidebands to a zero-bias Kondo peak have indeed been observed in the recent experiment by Yu *et al.*[5]. In contrast to the findings reported here, however, both satellite *peaks* and *dips* were observed in the nonlinear conductance.

We thank P. Brouwer, A. Rosch, P. Sharma and P. Wölfle for useful discussions. This research was supported by the Center for Functional Nanostructures (J. P.) and by the European Commission through project FP6-003673 CANEL of the IST Priority[19].

- 
- [1] M. A. Reed *et al.*, Science **278**, 252 (1997); R. H. M. Smit *et al.*, Nature **419**, 906 (2002); N. B. Zhitenev, H. Meng, and Z. Bao, Phys. Rev. Lett. **88**, 226801 (2002); S. Kubatkin *et al.*, Nature **425**, 698 (2003);
  - [2] H. Park *et al.*, Nature **407**, 57 (2000).
  - [3] J. Park *et al.*, Nature **417**, 722 (2002).
  - [4] W. Liang *et al.*, Nature **417**, 725 (2002).
  - [5] L. H. Yu and D. Natelson, Nano Lett. **4**, 79 (2004); L. H. Yu *et al.*, cond-mat/0408052.
  - [6] L. I. Glazman and R. I. Shekhter, Zh. Eksp. Teor. Fiz. **94**, 292 (1988) [Sov. Phys.-JETP **67**, 163 (1988)]; N. S. Wingreen, K. W. Jacobsen, and J. W. Wilkins, Phys. Rev. B **40**, 11834 (1989).
  - [7] K. Flensberg, Phys. Rev. B **68**, 205323 (2003).
  - [8] I. G. Lang and Y. A. Firsov, Sov. Phys. JETP **16**, 1301 (1963).
  - [9] H.-B. Schüttler and A. J. Fedro, Phys. Rev. B **38**, 9063 (1988).
  - [10] P. S. Cornaglia, D. R. Grempel, and H. Ness, cond-mat/0409021; cond-mat/0403578.
  - [11] Y. Meir and N. S. Wingreen, Phys. Rev. Lett. **68**, 2512 (1992).
  - [12] A. Rosch *et al.*, Phys. Rev. Lett. **90**, 76804 (2003); Phys. Rev. B **68**, 14430 (2003); cond-mat/0408506.
  - [13] J. König, H. Schoeller, and G. Schön, Phys. Rev. Lett. **76**, 1715 (1996).
  - [14] V. L. Gurevich and H. R. Schober, Phys. Rev. B **57**, 11295 (1998); K. R. Patton and M. R. Geller, Phys. Rev. B **67**, 155418 (2003).
  - [15] D. Boese and H. Schoeller, Europhys. Lett. **54**, 668 (2001); A. Mitra, I. Aleiner, and A. J. Millis, Phys. Rev. B **69**, 245302 (2004); M. Galperin, M. A. Ratner, and A. Nitzan, cond-mat/0405343.
  - [16] V. Aji, J. E. Moore, and C. M. Varma, cond-mat/0302222.
  - [17] A. Kogan, S. Amasha, and M. A. Kastner, Science **304**, 1293 (2004).
  - [18]  $f_{n'n} = \frac{e^{-\lambda^2/2}}{\sqrt{n'!n!}} [\text{sign}(n-n')\lambda]^{|n'-n|} L_{\min(n',n)}^{|n'-n|}(\lambda^2)$
  - [19] The views expressed in this publication are those of the authors and do not necessarily reflect the official European Commission's view on the subject.